

REMARKS

Claims 17-32 are on appeal to the Board of Appeals concerning the above-identified application.


In reply to the Notice of Non-Compliance applicants point out that the reference referred to was first cited by the Examiner, not by applicants. The reference throughout the prosecution of the application has been used together with Sugiyama et al in the rejection of the present claims based on 35 USC 103. Applicants were not the first to cite the reference in this application.

A replacement page 12 entitled EVIDENCE APPENDIX of the appeal brief is attached to this letter.

The application is believed to be in proper condition for consideration by the Board of Appeals.

Respectfully submitted,

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EVIDENCE APPENDIX

As to the matter of the citation of Japanese patent 05163342 in the record of the present application, appellants point out that the reference was first cited by the Examiner in the first Office Action of record in the case dated. July 3, 2007. The reference is listed in the Form 892 of the Office Action and was used, along with the Sugiyama et al patent, in the ground of rejection based on 35 USC 103. This ground of rejection was responded to in applicants' response filed September 14, 2007. Subsequently, this ground of rejection has been retained of record in the Office Actions of November 20, 2007, April 1, 2008 and November 10, 2008. Accordingly, it is the Examiner who has introduced the reference in question into the record. A copy of the cited reference, as originally received from the Patent Office, is enclosed for the Examiner's convenience.

Appellants enclose copies of the European Standard Test EN ISO 2440 for determining the compressive set of flexible slabstock foams under accelerated aging conditions. Also enclosed is an English language copy of inhouse test procedures identified as PB VWL 709 for determining the VOC and FOG values of condensable substances discharged from vehicle interior materials by thermodesorption.

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 (43)Date of publication of application : 29.06.1993  
 (21)Application number : 03-350813 (71)Applicant :ASAHI GLASS CO LTD  
 (22)Date of filing : 11.12.1991 (72)Inventor : TAKEYASU HIROMITSU  
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(54) PRODUCTION OF POLYETHER

(57)Abstract:

PURPOSE: To obtain a polyether of a controllable molecular weight and a controllable molecular structure by performing the ring opening addition reaction of a castor oil compound and a monoepoxide as an initiator in the persence of a compound metal cyanide complex catalyst.

CONSTITUTION: A polyether is produced by effecting the ring opening addition reaction of castor oil and/or modified castor oil with a 2C or higher monoepoxide as an initiator (e.g. ethylene oxide or propylene oxide) in the presence of a compound metal cyanide complex, desirably a complex having a structure of the formula, wherein M1 is Zn(II) or 2 like; M2 is Fe (II or III), Co (II or III) or the like; R is an organic ligand (e.g. ketone, ether, aldehyde, ester, alcohol or amide), and a, b, c, d, x and y are positive integers. According to the above process, a polyether of a freely controllable molecular weight and a freely controllable structure, for example, of an Mn of 2000-50000 and an Mw/Mn of 1.3 or above can be obtained.

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- 3.In the drawings, any words are not translated.

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 CLAIMS
 

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[Claim(s)]

[Claim 1]The manufacture approach of the polyethers characterized by carrying out the ring-opening addition reaction of the with a carbon numbers of two or more mono-epoxide by making castor oil and/or denaturation castor oil into an initiator under existence of a compound metallocyanide complex compound catalyst.

[Claim 2]The manufacture approach of the polyethers of claim 1 that the number average molecular weight of the polyethers obtained is 2000-50000.

[Claim 3]The manufacture approach of the polyethers of claim 1 that the weight average molecular weight/number average molecular weight of the polyethers obtained (Mw/Mn) are 1.3 or less.

[Claim 4]The manufacture approach of the polyethers of claim 1 that mono-epoxide is alkylene oxide.

[Claim 5]The manufacture approach of the polyethers of claim 4 that alkylene oxide is what is chosen from ethylene oxide and propylene oxide.

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 DETAILED DESCRIPTION
 

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[Detailed Description of the Invention]

[0001]

[Industrial Application]This invention relates to the manufacture approach of polyether polyol especially about the manufacture approach of polyethers.

[0002]

[Description of the Prior Art]Polyethers, such as polyoxyalkylene polyol which is made to carry out the ring opening reaction of the mono-epoxide, such as alkylene oxide, to an initiator, and is obtained, are widely used for the application of the raw material of synthetic resin, such as polyurethane, a surfactant, lubricant, and others. An initiator is an active hydrogen content compound expressed with A-(H) n (A: the residue except the hydrogen atom of an active hydrogen content compound, n:1 or more integers).

[0003]As an initiator, the compound which has hydroxy groups, such as monohydric alcohol, polyhydric alcohol, a univalent phenol, and a polyhydric phenol, for example, and the compounds (alkanolamines, amines-alkylene oxide addition product, etc.) which have a hydroxy alkylamino radical are mentioned. Moreover, the polyethers which mono-epoxide is made to react to the above-mentioned initiator, and are obtained are used as an initiator. Furthermore, the attempt using denaturation castor oil, such as castor oil and ester denaturation castor oil, as an initiator is also made.

[0004]Polyethers are the following compounds which are made to carry out the ring-opening addition reaction of the mono-epoxide to the above-mentioned initiator, and are obtained.

A[-(R-O) m-H] nA: Residue R-O except the hydrogen atom of an active-hydrogen content compound: The unit m in which mono-epoxide carried out ring breakage, n:1 or more integers [0005]Conventionally, the method of making mono-epoxide react to an initiator is widely used as an approach of manufacturing polyethers, under the alkali catalyst existence represented by alkali metal compounds, such as a potassium hydroxide and a sodium hydroxide. Moreover, in composition of the polyethers of low molecular weight, he is BF<sub>3</sub>. An acid catalyst like etherate is used.

[0006]

[Problem(s) to be Solved by the Invention]When a castor oil system compound was

used as an initiator, since decomposition of the unsaturated bond included in a castor oil system compound or an ester bond was produced, the alkali catalyst known conventionally was not able to be used. When the acid catalyst of Lewis acid etc. was used, the amount polyethers of macromolecules were hard to be obtained.

[0007]

[Means for Solving the Problem] This invention is the manufacture approach of the polyethers of the amount of comparison-macromolecules which was made that the above-mentioned trouble should be solved and made especially the castor oil system compound the initiator. That is, it is the manufacture approach of the polyethers characterized by carrying out the ring-opening addition reaction of the with a carbon numbers of two or more mono-epoxide by making castor oil and/or denaturation castor oil into an initiator under existence of a compound metallocyanide complex compound catalyst.

[0008] The castor oil and denaturation castor oil which are used by this invention are denaturation castor oil obtained by processing an ester exchange reaction, a hydrogenation reaction, a sodium reduction reaction, etc. in the natural oil and this natural oil which are obtained from the seed of a castor seed.

[0009] Castor oil is partial saturation ester which consists of a glyceride of the partial saturation acid which uses a ricinoleic acid as a principal component, and has a hydroxyl group. It is possible to control the number of functional groups freely by an ester exchange reaction etc.

[0010] Especially the with a carbon numbers of two or more mono-epoxide made to react to castor oil and/or denaturation castor oil in this invention has with a carbon numbers of three or more desirable alkylene oxide. Still more preferably, the alkylene oxide of the carbon numbers 3-4, such as ethylene oxide, propylene oxide, 1, 2-butylen oxide, 2, 3-butylen oxide, and epichlorohydrin, is desirable, and is ethylene oxide and propylene oxide most preferably.

[0011] These independent mono-epoxide of others, such as those two or more sorts or they and styrene oxide, glycidyl ether, and glycidyl ester, can be used together and used. In use of two or more sorts of alkylene oxide, or use of alkylene oxide and other mono-epoxide, they can be mixed and added, or sequential addition can be carried out, and a random polymerization chain and a block polymerization chain can be formed.

[0012] Manufacturing polyethers using the compound metallocyanide complex of this invention is known (USP.3278457, USP.3278458, USP.3278459 specification). This catalyst has little generation of partial saturation mono-ox, and it is also possible to manufacture the polyethers of the amount of macromolecules extremely.

[0013] It is thought that the compound metallocyanide complex in this invention has the structure of the following general formula (1) as shown in the above-mentioned well-known example.

$M1a[M2x(CN)y]b(H_2O)cRd \quad (1)$

However, M1 Zn (II), Fe (II), Fe (III), Co (II), nickel (II), aluminum (III) and Sr (II), Mn (II), Cr (III), Cu (II), Sn (II), Pb (II), Mo (IV), Mo (VI), It is W (IV), W (VI), etc. and is M2. Fe (II), Fe (III), Co (II), Co (III) and Cr (II), Cr (III), Mn (II), It is Mn (III), nickel (II), V (IV), V (V), etc., R is an organic ligand, a, bx, and y are positive integers which change with a metal valence and the metal coordination number, and c and d are positive numbers which change with the coordination number to a metal.

[0014] M1 in a general formula (1) Zn (II) is desirable and it is M2. Fe (II), Fe (III) and Co (II), Co (III), etc. are desirable. As an organic ligand, there are a ketone, the ether, an aldehyde, ester, alcohol, an amide, etc., for example.

[0015] The compound metallocyanide complex expressed with a general formula (1) is metal salt M1 Xa (anion in which, as for M1 and a, X forms M1 and salt like \*\*\*\*), and poly cyano meta rate (salt) Ze  $[M2x(CN)y]f$  (M2, x, and y are the

same as that of \*\*\*\*.). Z is hydrogen, alkali metal, alkaline earth metal, etc. e and f are Z and M2. After mixing the solution of the mixed solvent of each water solution or water of the positive integer decided by the valence and the coordination number, and an organic solvent and contacting an organic ligand R to the obtained compound metallocyanide, it is manufactured by removing an excessive solvent and an excessive organic ligand R.

[0016]The poly cyano meta rate Ze (salt)  $[M2x(CN) y] f$  Although various metals including hydrogen or alkali metal can be used for Z, lithium salt, sodium salt, potassium salt, magnesium salt, and a calcium salt are desirable. It is the especially desirable usual alkali-metal salt, i.e., sodium salt and potassium salt.

[0017]The polyethers which have a hydroxyl group are used with the poly isocyanate compound as a raw material for polyurethane manufacture. The polyethers which have this hydroxyl group add mono-epoxide, especially alkylene oxide to the initiator which has a polyhydroxy compound, an amine compound, and other at least one active hydrogen, and it is manufactured.

[0018]In this addition reaction, alkali-metal hydroxides, such as a potassium hydroxide and a sodium hydroxide, are most ordinarily used as a catalyst. Use is also proposed for catalysts, such as 3 boron fluoride and tertiary amine, by the pan.

[0019]However, when the addition reaction of the mono-epoxide was carried out by making a castor oil system compound into an initiator, there were problems, like the molecular weight distribution of the obtained polyethers from which the polyethers of the amount of macromolecules will be hard to be obtained if there is a trouble that a castor oil system compound decomposes when an alkali catalyst is used, and the acid catalyst of Lewis acid etc. is used become large.

[0020]Since a compound metallocyanide complex compound catalyst is used for this invention, it can add mono-epoxide by making castor oil and /denaturation castor oil into an initiator. Moreover, control of molecular weight is possible and it is also possible to manufacture the polyethers of the amount of macromolecules comparatively.

[0021]Although the approach of using the metalloporphyrin other than the approach using a compound metallocyanide complex compound catalyst as an approach of compounding the polyethers of the amount of macromolecules comparatively, without using alkali or an acid catalyst using the usual initiator is learned (JP,61-197631,A), when polyethers are manufactured using metalloporphyrin, there is a problem of the polyethers obtained coloring.

[0022]When the polyethers which have the hydroxyl group manufactured by this invention are used as a polyurethane raw material, it may have a bad influence on the physical properties of the polyurethane reacted or generated at the time of the catalyst which remains in polyethers being polyurethane manufacture. It is desirable after polyethers manufacture to remove a residual catalyst.

[0023]In order to remove this catalyst from the polyethers using a compound metallocyanide complex compound catalyst, it is required to decompose and ionize a catalyst from alkali or an acid it not only to to process with filtration, an adsorbent, etc., but, and to remove these decomposition products, residual alkali, and a residual acid by adsorption, filtration, or extract after that.

[0024]The approach (JP,2-289617,A, JP,2-289618,A) of using together the approach (JP,59-15336,B) of decomposing with alkali metal or an alkali metal compound, the approach (JP,2-276821,A) of decomposing by the alkali-metal alcoholate, a buffer for pH or a buffer for pH, and a chelating agent as the approach of decomposition of a catalyst and removal, and decomposing and removing is found out. In order to remove a catalyst from the polyethers of this invention, the method of using a buffer for pH is suitable.

[0025]As for the number average molecular weight of the polyethers which have the hydroxyl group obtained in this invention, 2000-50000 are desirable. Especially

2000-30000 are desirable.

[0026] Moreover, as for the polyethers which have the hydroxyl group obtained in this invention, it is desirable that the value of weight average molecular weight/number average molecular weight (referred to as Mw/Mn below) is small. They are especially Mw/Mn. 1.3 or less are desirable.

[0027] Although the castor oil and denaturation castor oil of this invention have a hydroxyl group and it has a carboxyl group further by the case, if a compound metalocyanide complex compound catalyst is used, it is possible to add mono-epoxide to both this functional group. And the obtained polyethers have only a hydroxyl-group end.

X [0028] The polyethers which have the hydroxyl group obtained by this invention are ~~the most useful as polyol for polyurethane raw materials~~ which is independent [ its ], or uses together with other polyols and is used. Moreover, the polyethers obtained by this invention are used also for the raw material of synthetic resin other than polyurethane, or the application of an additive. Furthermore, it can use as a lubricating oil, insulating oil, hydraulic oil, other oils, or its raw material. Furthermore, the polyethers obtained by this invention are changed into other compounds, such as an alkyl ether ghost and an acylation object, and it can be used for various applications.

[0029]

[Example] The addition reaction of alkylene oxide was performed to the initiator of the following castor oil system using each catalyst.

Initiator A: Polyol of three functional groups denaturalized and obtained in castor oil, and a hydroxyl value 160 (product made from Ito Oil Mill URIC H-30)

Initiator B: Polyol of three functional groups denaturalized and obtained in castor oil, and a hydroxyl value 90 (product made from Ito Oil Mill URIC H-57)

Initiator C: Polyol of four functional groups denaturalized and obtained in castor oil, and a hydroxyl value 320 (product made from Ito Oil Mill URIC H-91)

[0030] [Example 1] 200 ppm (opposite workmanship) of zinc hexa cyano cobaltate catalysts were used for the 1000g initiator A, propylene oxide 1920g was added under 110-degree C conditions, and polyether polyol was obtained.

[0031] [Example 2] 253 ppm (opposite workmanship) of zinc hexa cyano cobaltate catalysts were used for the 1000g initiator B, after processing with ammonia the polyol which the 70/30wt% mixture of propylene oxide and ethylene oxide was made to react under 105-degree C conditions, and was obtained [ mixture ] in 1700g, with the synthetic magnesium silicate, adsorption treatment was carried out, it filtered, and polyether polyol was obtained.

[0032] [Example 3] After using 250 ppm (opposite workmanship) for the 1000g initiator C for the zinc hexa cyano cobaltate catalyst and processing with ammonia propylene oxide 1000g and the polyol which the 40/60wt% mixture of propylene oxide/ethylene oxide was made to react under conditions (3800g and 110 degrees C) continuously, and was obtained, with the synthetic magnesium silicate, adsorption treatment was carried out, it filtered, and polyether polyol was obtained.

[0033] [Example 4] 2000 ppm (opposite workmanship) were taught by having made the potassium hydroxide into the catalyst at the 1000g initiator A, the same reaction as Example 1 was performed, the synthetic magnesium silicate performed adsorption treatment, and polyether polyol was obtained.

[0034] [Example 5] He is BF<sub>3</sub> to the 1000g initiator B. 2000 ppm (opposite workmanship) of etherate were taught, the same reaction as Example 2 was performed, the synthetic magnesium silicate performed adsorption treatment of a catalyst, and polyether polyol was obtained.

[0035] [Example 6] 3000 ppm (opposite workmanship) were taught by having made the potassium hydroxide into the catalyst at the 1000g initiator C, the same reaction as Example 3 was performed, adsorption treatment of a catalyst was performed

using the synthetic magnesium silicate, and polyether polyol was obtained. the description of the polyether polyol obtained in Examples 1-6 -- a value is shown in Table 1.

[0036]

[Table 1]

		外 観	水酸基価	$M_n / M_0$
実 施 例	例1	透 明	56.3	1.10
	例2	透 明	33.2	1.12
	例3	透 明	56.0	1.13
比 較 例	例4	茶 褐 色	82.5	1.7
	例5	黒色に着色	80.0	1.8
	例6	茶 褐 色	70.2	1.75

[0037]

[Effect of the Invention] This invention is the approach of manufacturing the polyethers which made the ~~castor oil system compound the initiator~~. Control of the conventionally impossible molecular weight or structure can be performed freely.